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TITLE. MEASUREMENT OF RADIOACTIVE CONTAMINATED WASTES

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MEASUREMENT OF RADIOACTIVE CONTAMINATED WASTES

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At Los Alamos, a comprehensive program is underway for the development of sensitive, practical, nondestructive assay techniques for the quantification of low-level transuranics in bulk solid wastes. The program encompasses a broad range of techniques, including sophisticated active and passive gamma-ray spectroscopy, passive neutron detection systems, pulsed portable neutron generator interrogation systems, and electron accelerator-based techniques. The techniques can be used with either low-level or high-level beta-gamma wastes in either low-density or high-density matrices. The techniques are quite sensitive (<10 nCi/g detection) and, in many cases, isotopic specific. Waste packages range in size from small cardboard boxes to large metal or wooden crates. Considerable effort is being expended on waste matrix identification to improve assay accuracy.

INTRODUCTION

As part of any comprehensive radioactive waste management program, it is important to have the capability to determine the types and amounts of radioactivity in wastes. Without such ability, it is impossible to direct cost effective and timely waste management programs for waste generators and repositories, the decontamination and decommissioning of outdated nuclear facilities, and the exhumation of old radioactive waste burial grounds. Heretofore, only administrative controls could be used for transuranic (TRU) waste sorting, often resulting in overestimates of TRU waste quantity, thereby significantly increasing the amount of materials that had to be handled as retrievable wastes, which requires expensive handling techniques and engineered storage.

To ameliorate this problem, a comprehensive program is in progress at the Los Alamos National Laboratory for the development of sensitive, practical, nondestructive assay techniques for the quantification of low-level transuranics in bulk solid wastes. The program encompasses a broad range of techniques, including sophisticated active and passive gamma-ray interrogation, passive neutron detection systems, pulsed portable neutron generator interrogation systems, and electron accelerator-based techniques. The techniques can be used with either low-level or high-level beta-gamma wastes in either low-density or high-density matrices. Waste package containers range in size from 2 ft^3 cardboard cartons to 4x4x7' wood and metal crates. Through the use of such techniques, the capability now exists to cost-effectively sort and segregate wastes by TRU content. The techniques are generic in nature and can be effectively applied to other than TRU wastes, such as uranium and thorium mill tailings and radiopharmaceutical wastes.

COMBUSTIBLE WASTE ASSAY

One of the first instruments designed and built for the assay of transuranic wastes at the 10 nCi/g level was the Multienergy Gamma Assay System (MEGAS).[1] The original MEGAS (see Fig. 1) has been significantly upgraded (MEGAS II).[2-4] MEGAS II operates in a segmented mode, which allows the determination of hot spots within waste packages. The basic photon detector for both is a 127-mm diameter by 1.6-mm thick NaI crystal, which optimizes the TRU detection capability using L x rays and gamma rays having an energy less than 100 keV. The detection limit at the 3σ level above background for ^{241}Am is less than 5 pCi/g and for ^{239}Pu is less than 1 nCi/g for a 500-s count. Packages typically contain low density, combustible type wastes in a 2 ft^3 cardboard carton. Total mass of the container and wastes must generally be less than 10 kg for accurate assay (errors ±50%).

Fig. 1. MEGAS TRU waste assay system with NaI photon detector on top of support looking at rotating waste package.

The presence of beta- and gamma-ray emitting fission products decreases the TRU detection limit for the NaI detector. The addition[3,4] of a high resolution, hyperpure, planar germanium detector, 1000-mm^2 active area, 12-mm thick, allows the assay of TRU isotopes even in the presence of several mR/h gamma and beta backgrounds. A tabulation of measured detection limits for the hyperpure germanium detector is presented in Ref. 3. Using these data, it is estimated that TRU assay at the 10 nCi/g level can still be made even in the presence of 400 μCi of ^{137}Cs (65 nCi ^{137}Cs/g).

When four banks of polyethylene-moderated ^3He detectors are added[3,4] around the MEGAS, neutron detection is achieved. The measured detectability limit

(3σ level above background, 1000-s count, total neutron count) for these neutron detectors is 400 nCi/g for defense grade plutonium oxide. Because ^{3}He detectors are relatively insensitive to photons, they can operate even in the presence of high fission product backgrounds (1-10 R/h). The neutron counters were added to assure that significant quantities (>100 mg) of plutonium, even if well shielded, would be detected. The technology for this development has been transferred to other DOE facilities and to the U.S. commercial instrumentation sector.

CRATE COUNTER

Much of the plutonium and uranium waste generated in the nuclear industry is ultimately packaged in large crates having typical dimensions of 1.0-m or more on a side. An active-passive 4π neutron counting system has been developed at Los Alamos to assay/screen these large containers for their TRU and uranium content. This crate counter is made from discrete moderated ^{3}He neutron detector modules which are easily arranged into a variety of assay chamber geometries. Very large objects and debris from decommissioning programs can be easily accommodated in the counter.

Fig. 2. Schematic of modular 4π neutron assay chamber for large crate assay

Figure 2 depicts the construction of the discrete counter modules and the placement to form an assay chamber with internal dimensions of 1.2x1.2x2.4 m. The measured 4π detection efficiency in this configuration for a bare californium source is 14%. Separate counting electronics are provided for each of the two chambers in each of the six modules for a total of twelve independent signals. The relative singles count rates from different portions of the 4π system are used for geometry and matrix corrections. Figure 3 shows the ratio of the count rates from the two end modules as a function of source position along the length of the assay chamber. Similar ratios have been measured for the two sets of opposite side modules.[5] The three ratios can serve to determine the approximate location of a source. Figure 4 shows the crate counter in final assembly. This system will be placed in routine operation at a DOE facility.

All neutron detection systems suffer in the presence of matrix materials, particularly hydrogenous materials. A flat response (±10%) was measured for ^{252}Cf neutron sources moderated by thicknesses of polyethylene ranging from 0 to 7.5 cm. A compensation technique for greater effective hydrogenous moderators is based on the differential energy sensitivity of the

Fig. 3. Ratio of count rates for the end modules of a 4π neutron crate assay chamber for a source moved along the length of the assay chamber.

Fig. 4. TRU crate assay system in final construction phase.

count rates in the inner and outer chambers of each module.

For plutonium contaminated wastes, the passive 4π coincident measurement generally determines the ^{240}Pu mass. If the ^{240}Pu to total Pu ratio is known, this measurement determines the total Pu mass. Measured 3σ detection sensitivity is about 10 mg ^{240}Pu.[5,6]

The active part of the crate counter is the differential die-away pulsed neutron technique discussed next and elsewhere.[7] Preliminary detection limits for the active crate counter are 5-10 mg for either ^{239}Pu or ^{235}U.[6]

55-GALLON DRUM COUNTER

Los Alamos has also developed an accurate, highly sensitive assay system for the measurement of TRU waste in 208-ℓ (55-gallon) barrels. The assay chamber of this differential die-away system (see Fig. 5) consists of a graphite and polyethylene structure with a small, pulsed D + T neutron generator inside. Both cadmium-covered and bare ^{3}He neutron detectors are incorporated in the chamber, being placed external to the graphite but within polyethylene. The graphite and polyethylene moderated 14 MeV neutrons are completely thermalized in 0.7 ms. The thermal neutron

Fig. 5. Schematic of drum assay system.

die away in the interrogation cavity with a half life of about 0.76 ms. The interrogating thermal pulse lasts a long time in the chamber and induces thermal neutron fission in any fissile material present in the waste barrel. The cadmium-covered detectors count only the fast, fission-produced neutrons. This system has a measured ^{239}Pu sensitivity of less than 1 mg in a 208-ℓ barrel. A complete description of this system, including its application to mixed wastes (curium, californium, plutonium, uranium, americium, neptunium), and matrix correction methods, is presented elsewhere.[8,9] A drum assay system has recently been developed and fabricated by Los Alamos and installed at the Oak Ridge National Laboratory for test and evaluation purposes under field conditions (see Fig. 6). The system is working as planned.

Fig. 6. ORNL drum assay system with door open and barrel loading platform in place.

GAMMA ASSAY

An ideal supplement to the pulsed thermal neutron interrogation system is gamma-ray spectroscopy. This is particularly true for waste containing many isotopes. Gamma-ray spectroscopy is sensitive to most radioactive isotopes, notably fission products, and many TRU isotopes that are difficult to assay by active or passive neutron methods. Some specific examples of isotopes that cannot be assayed via neutrons that can be assayed using gamma-ray spectroscopy are ^{243}Am and ^{237}Np, which have daughters, ^{239}Np and ^{233}Pa, respectively, that have energetic gamma rays. An isotope in the grey area is ^{241}Am, which has a low fission cross section and spontaneous fission rate and a very intense but low energy (E_γ = 60 keV), gamma ray.

Active/passive gamma-ray spectroscopy has been long used to quantify the radioactive material and wastes in a barrel.[10,11] The major problem is characterizing the matrix to make the necessary corrections to the gamma-ray signatures. There are two subtly different techniques. One is to use external sources, identical to the isotopes in the barrel, to over-ride the passive signal, to give the effective attenuation at the desired energies. The other technique relies on the fact that above about 150 keV, the attenuation coefficient varies smoothly and slowly as a function of energy. This technique characterizes the matrix as a function of energy over a large energy range.

LINAC APPLICATIONS

An electron linear accelerator (LINAC) can be the heart of a complete assay system. Photofission interrogation offers good sensitivity for TRU, but because of the similarity of photofission cross sections for both fissile and fertile (e.g. ^{238}U) isotopes and other high Z materials, such as lead, identification of specific nuclides is difficult. Thermal neutron interrogation offers high sensitivity for fissile isotopes but essentially none for fertile isotopes. A combination of neutron and photon interrogation can separate the fissile and fertile isotopes.[12]

Photons are produced in a bremsstrahlung target that stops the electron beam. The photon beam then passes through a polyethylene slab to harden the photon spectrum. A portion of the higher energy photons above various reaction threshold energies will produce photoneutrons. A beryllium converter can also be used to significantly increase the photoneutron flux. Photoneutrons and prompt photofission neutrons will thermalize in a few tens of microseconds and will persist as thermals for hundreds of microseconds, during which time they will generate thermal neutron fissions among the fissile TRU. Prompt fissions from thermal fission are separated in time from the photoneutrons and can serve as a quantitative signature. The detection method is the differential die-away system described earlier and elsewhere.[7-9]

Fig. 7. Neutron count rate vs. time from simultaneous photon and neutron interrogation of 1 g ^{239}Pu.

While the thermal fissions are produced in near simultaneity with the photofission events, the prompt and delayed neutrons from the two fission processes can be resolved in a single detector. This is illustrated in Fig. 7, where the neutron count rate from a 1 g ^{239}Pu sample irradiated by a 12 MeV bremsstrahlung burst is plotted versus time after burst. Curve "a" shows the prompt neutron count rate to persist for about 8 ms, with a nearly constant delayed neutron count rate continuing to the next burst. Curve "b", obtained with the ^{239}Pu wrapped with cadmium, shows the delayed neutrons to be only weakly affected by the cadmium, whereas the prompt neutrons are essentially absent, demonstrating the predominantly photofission origin of the delayed group. For a 200-s LINAC interrogation run, the 3σ detection limit is better than 1 mg ^{239}Pu, which is less than 1 nCi/g of waste for a 105 kg matrix of aluminim scrap in a 208-ℓ barrel.[13,14] Barrels of concrete, bitumen, sand, and other matrices have also been studied with wide applications in waste management programs. It should be noted that the systems described here can be installed in a mobile van for field analysis. Such a system is now under study.

NONDESTRUCTIVE EXAMINATION

While the LINAC is being used as an interrogation source, it can simultaneously be used to produce a radiograph or picture of the waste container and contents. A radiograph indicates where and what inhomogeneities are in the barrel. Of a purely qualitative nature, a radiograph gives an excellent fingerprint of the barrel, which can be used for shipper/receiver verification that a given barrel has not been tampered with.

MATRIX IDENTIFICATION

To further complete an assay, the LINAC can be used to identify matrix constituents using the thermal neutron capture reaction (n,γ) and a hyperpure germanium (HpGe) gamma detector. Such identification is especially useful when the neutron detectors indicate highly absorbing materials present. Preliminary measurements have identified cadmium, aluminum, iron, hydrogen and chlorine. In our preliminary measurements, the HpGe detector was severely affected by the gamma flash from the LINAC and was paralyzed for several milliseconds after the flash. Thus, the prompt capture gamma rays were missed and only a few gamma rays from thermal neutron activation were detected. Recent efforts have greatly reduced the paralysis time of the detector so that prompt capture gamma rays are now detected.

Similar matrix studies can be made using other external neutron sources, such as ^{252}Cf, or even the internal neutron sources (e.g. alpha-n reactions) contained within the waste. Generally, these methods do not suffer from detector paralysis problems. Table I shows the experimentally measured detectability limit (3σ level above background, 1000-s count) for various elements located in the central region of a 208-ℓ barrel (~100 kg). These measurements employed a 50 μg ^{252}Cf neutron source and a highly collimated 16% efficient Ge(Li) detector. Thermal neutron capture is particularly sensitive to neutron poisons. A detectability limit below 1 g (~10 ppm) is achieved for all the neutron poisons except lithium. The poor sensitivity for lithium is because the primary neutron absorbing lithium isotope, ^{6}Li, captures neutrons without emitting gamma rays. The technique can be used to identify hazardous and toxic materials other than radioactive materials (e.g. the heavy metals, mercury and cadmium).

TABLE I. Elemental Thermal Neutron Capture Gamma-Ray Sensitivities for 208-Liter-Drum Assays.

Element	Number of Gamma-Ray Lines[a]	Detectability Limit[b]
Hydrogen[c]	1	14.2 g
Helium	0	
Lithium[d]	7	3.2 kg
Beryllium	7	8.6 kg
Boron[d]	7	150 mg
Carbon	3	40.3 kg
Nitrogen[c]	43	1.7 kg
Oxygen	0	
Fluorine	11	5.6 kg
Sodium	51	176 g
Magnesium	18	286 g
Aluminum[c]	51	605 g
Silicon	27	970 g
Phosphorus	60	2 kg
Sulfur	33	400 g
Chlorine	41	15.3 g
Potassium	88	280 g
Calcium	46	792 g
Scandium	87	21.8 g
Titanium	39	45.2 g
Vanadium	62	68.2 g
Chromium	56	202
Manganese	76	48.2 g
Iron[c]	42	508 g
Cobalt	59	29 g
Nickel	49	122 g
Copper	66	95 g
Zinc	71	1.2 kg
Cadmium[d]	33	420 mg
Gadolinium[d]	17	879 mg
Mercury	41	3.2 g

[a]When usable, escape peaks are included.

[b]Counting time 1000-s, three standard deviations above background.

[c]Possible interference with measurement system components.

[d]Neutron poison.

SUMMARY

The Los Alamos TRU waste assay program is developing measurement techniques for TRU and other radioactive waste materials generated by the nuclear industry. Systems are now being fielded for test and evaluation purposes at DOE TRU waste generators. The transfer of this technology to other facilities and the commercial instrumentation sector is well in progress.

REFERENCES

1. C. J. Umbarger and L. R. Cowder, "Measurement of Transuranic Solid Wastes at the 10 nCi/g Activity Level," Nucl. Tech 27, 500 (1975).

2. J. S. Hendricks and D. A. Close, "^{3}He Detector Design for Low Level Transuranic Waste Assay," Trans. ANS 28, 137 (1978).

3. D. A. Close, et al., "TRU Waste Assay Instrumentation: New Developments and Directions at the Los Alamos Scientific Laboratory," Journal of the INMM 7, 503 (1978).

4. T. W. Crane, L. R. Cowder, D. F. Jones, and E. R. Martin, "Assay of Fission Product Contaminated Waste at the 10 nCi/g Fiducial," Journal of the INMM 6, 446 (1977).

5. E. R. Shunk, J. T. Caldwell, S. W. France, and G. C. Herrera, "A High-Efficiency, Modular System for the Assay of Plutonium in Large Waste Packages, presented at the IEEE Nuclear Science Symposium, San Francisco, California, October 21-23, 1981; Los Alamos National Laboratory Report LA-UR-81-3078 (1981).

6. J. T. Caldwell, "Passive and Active Assay Measurements on Crate Size Waste Packages," Trans. ANS 39, 339 (1981).

7. W. E. Kunz, J. D. Atencio, W. Bernard, G. C. Herrera, J. C. Pratt, and J. T. Caldwell, "A 1-mg Sensitivity Fissile Assay System," Proc. 3rd ESARDA Symposium, Karlsruhe, F. R. Germany, May 6-8, 1981, p. 119.

8. W. E. Kunz, J. D. Atencio, W. Bernard, G. C. Herrera, H. H. Hsu, J. C. Pratt, and J. T. Caldwell, "Multi-Isotopic Waste Assay System," ANS Topical Meeting on Treatment and Handling of Radioactive Wastes," Richland, Washington, April 19-22, 1982.

9. J. T. Caldwell, W. E. Kunz, J. D. Atencio, G. C. Herrera, and J. C. Pratt, "Experimental Evaluation of the Differential Die-away Pulsed Neutron Technique for the Fissile Assay of Hot Irradiated Fuel Wastes," ANS Topical Meeting on Treatment and Handling of Radioactive Wastes," Richland, Washington, April 19-22, 1982.

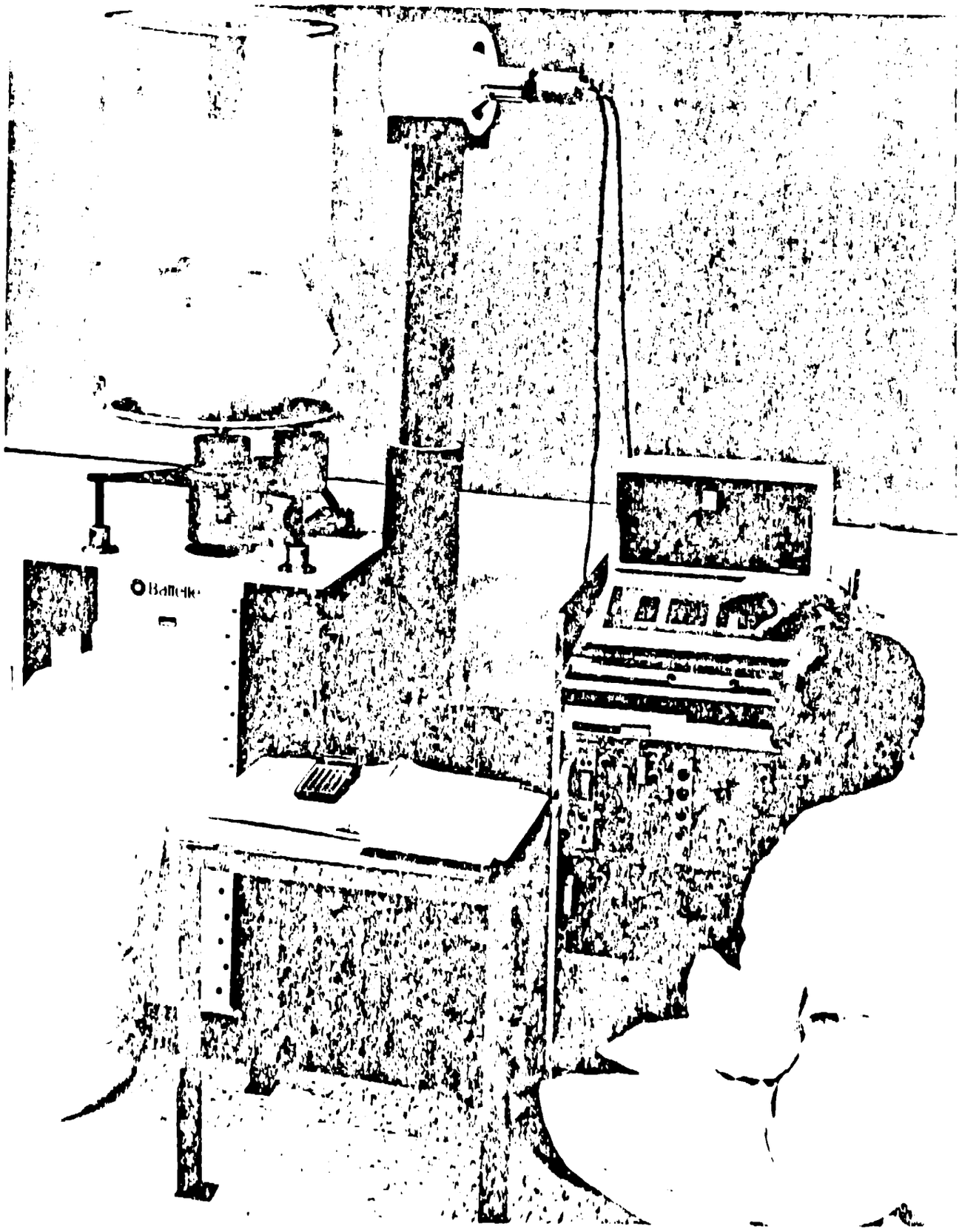
10. J. L. Parker, T. D. Reilly, R. B. Walton, D. B. Smith, and L. V. East, "Attenuation Corrected Passive Gamma Assay," Los Alamos Scientific Laboratory Report LA-4705-MS (1970).

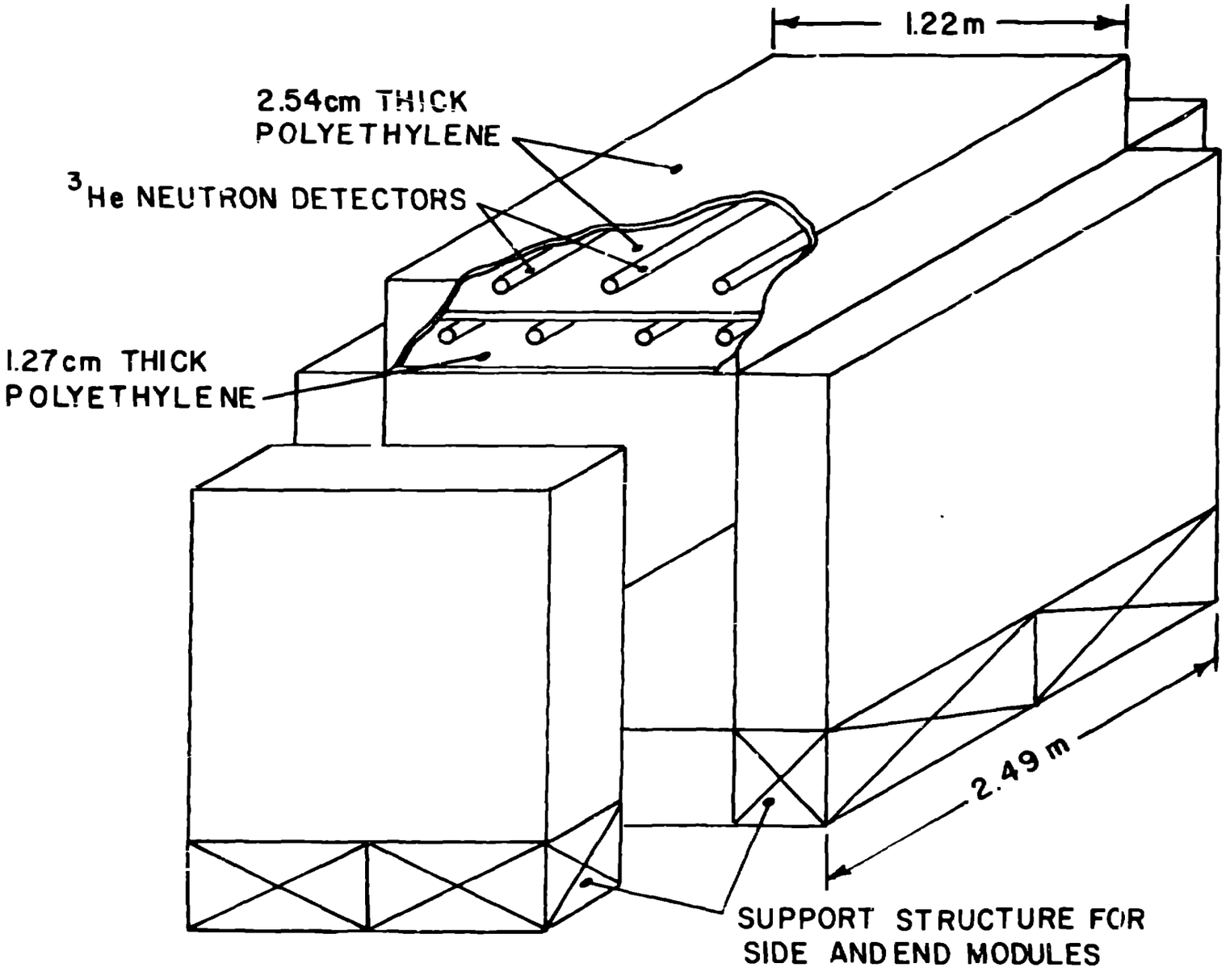
11. L. V. East, E. R. Martin, T. L. Atwell, R. H. Augustson, and H. O. Menlove, "Automated Nondestructive Assay Instrumentation for Nuclear Materials Safeguards," IEEE Trans. Nucl. Sci. 22, 739 (1975).

12. L. A. Franks, J. L. Pigg, J. T. Caldwell, M. R. Cates, W. E. Kunz, B. W. Noel, and D. A. Close, "High Sensitivity Transuranic Waste Assay by Simultaneous Photon and Thermal-Neutron Interrogation Using an Electron Linear Accelerator," Nucl. Inst. and Meth., 193, 571 (1982).

13. J. T. Caldwell, M. R. Cates, D. A. Close, T. W. Crane, W. E. Kunz, E. R. Shunk, C. J. Umbarger, and L. A. Franks, "Recent Developments at Los Alamos for Measuring Alpha-Contaminated Waste," International Symposium on the Management of Alpha-Contaminated Wastes, Vienna, Austria, June 2-6, 1980, IAEA-SM-246/67.

14. M. R. Cates, B. W. Noel, J. T. Caldwell, W. E. Kunz, D. A. Close, L. A. Franks, and J. L. Pigg, "Application of Linear Accelerator Technology to the Detection of Trace Amounts of Transuranics in Waste Barrels," International Symposium on the Management of Alpha-Contaminated Wastes, Vienna, Austria, June 2-6, 1980, IAEA-SM-246/30.




1.22m
2.54cm THICK
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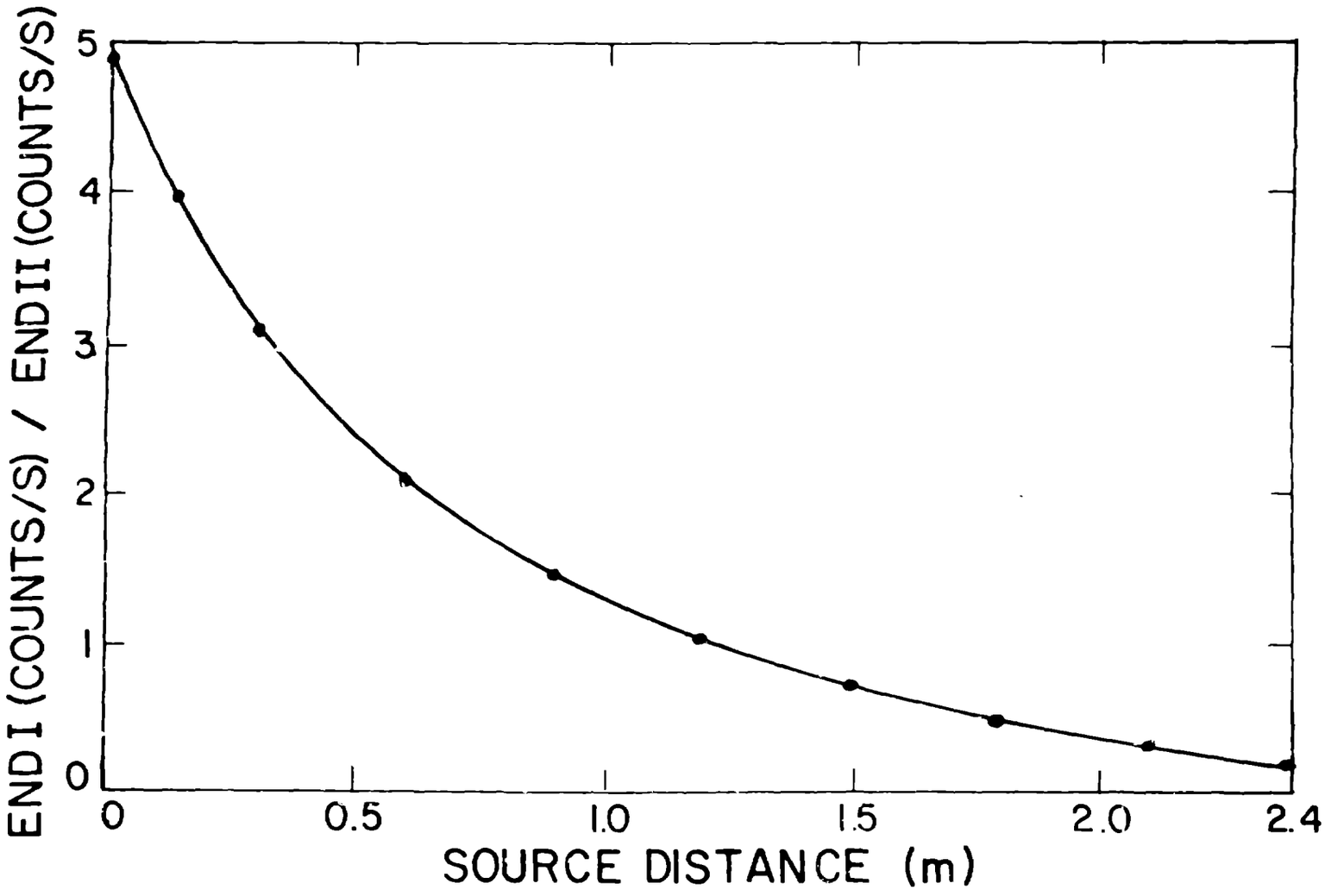
2.49 m
SUPPORT STRUCTURE FOR
SIDE AND END MODULES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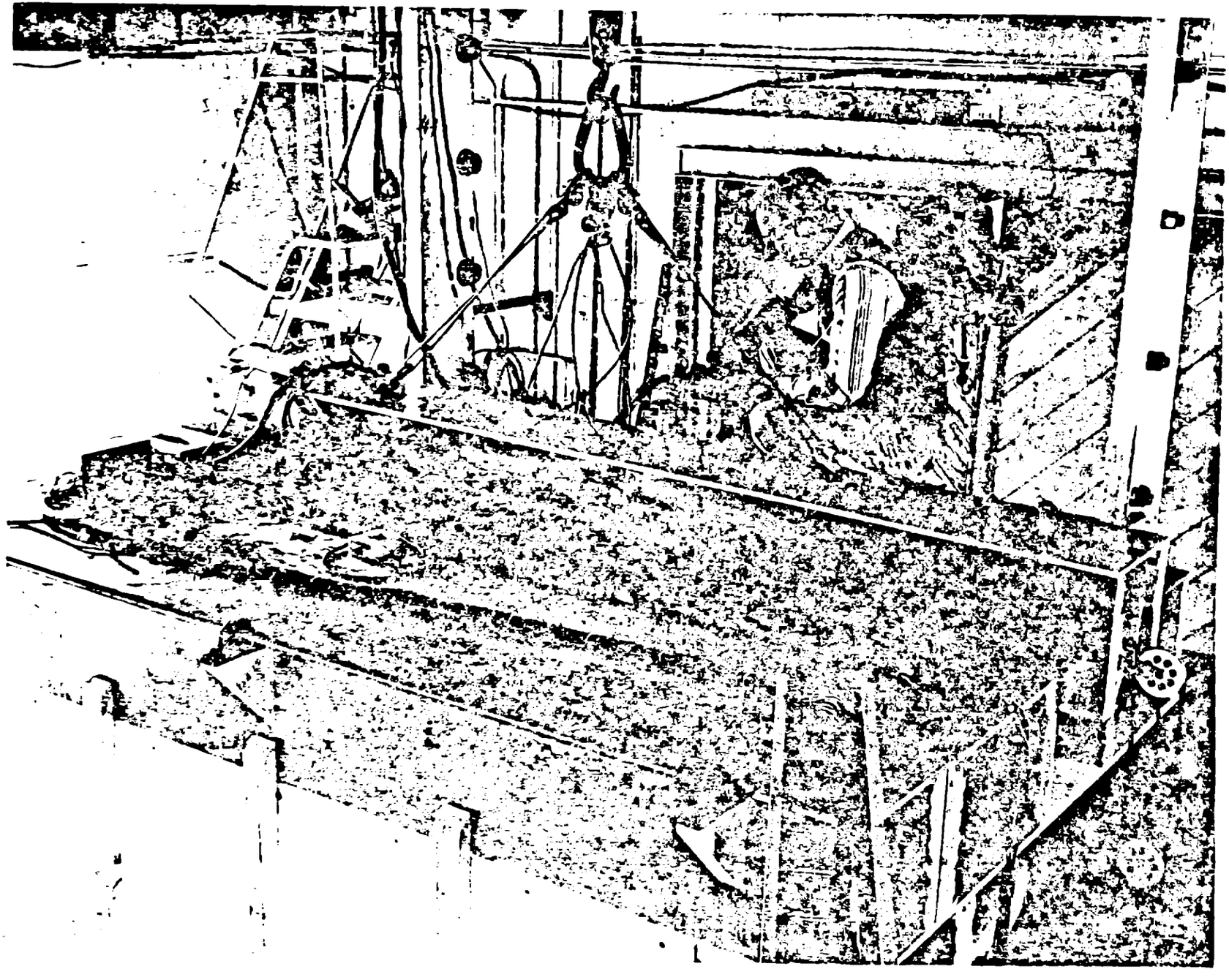

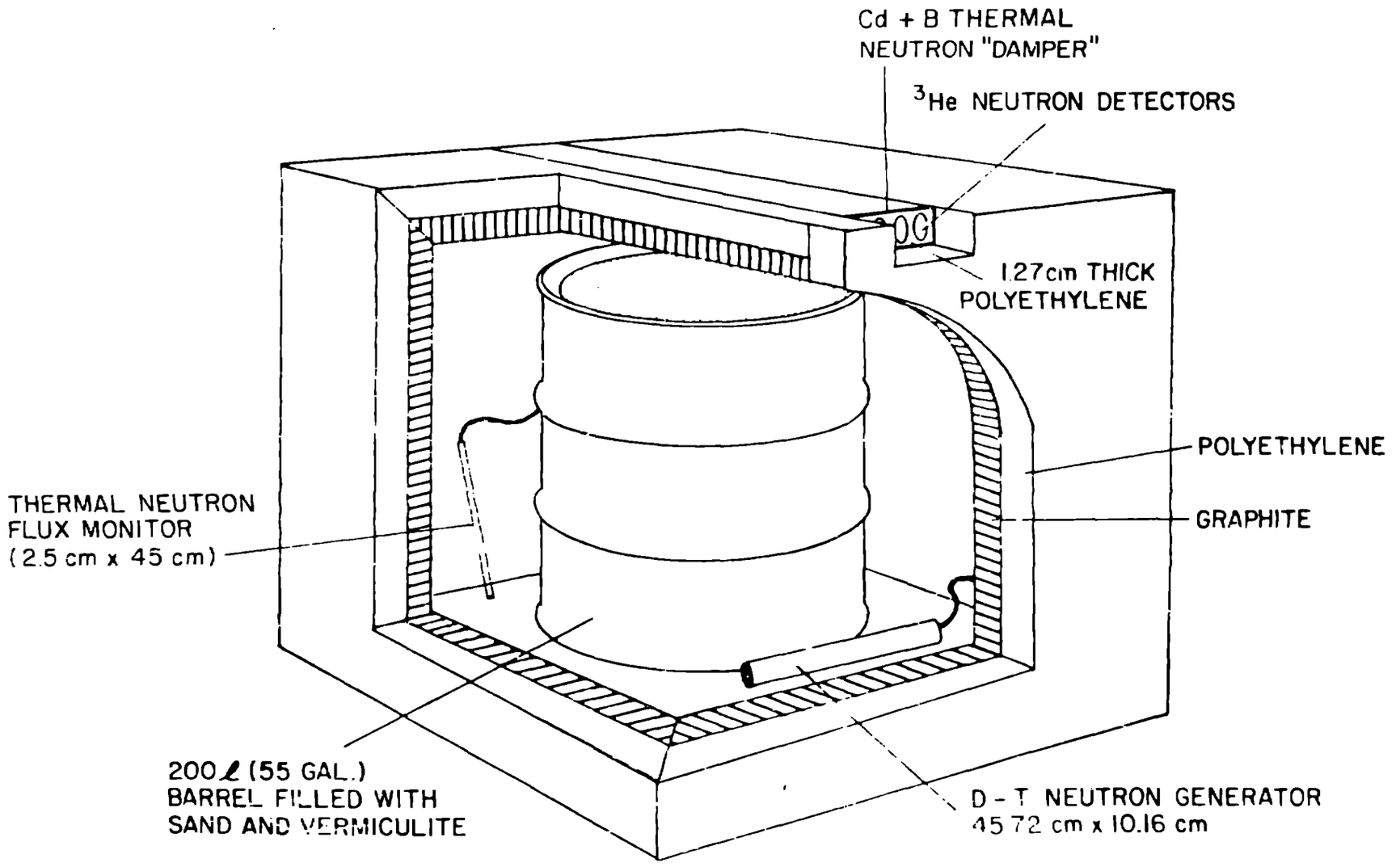
Cd + B THERMAL
NEUTRON "DAMPER"
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POLYETHYLENE
POLYETHYLENE
GRAPHITE
THERMAL NEUTRON
FLUX MONITOR
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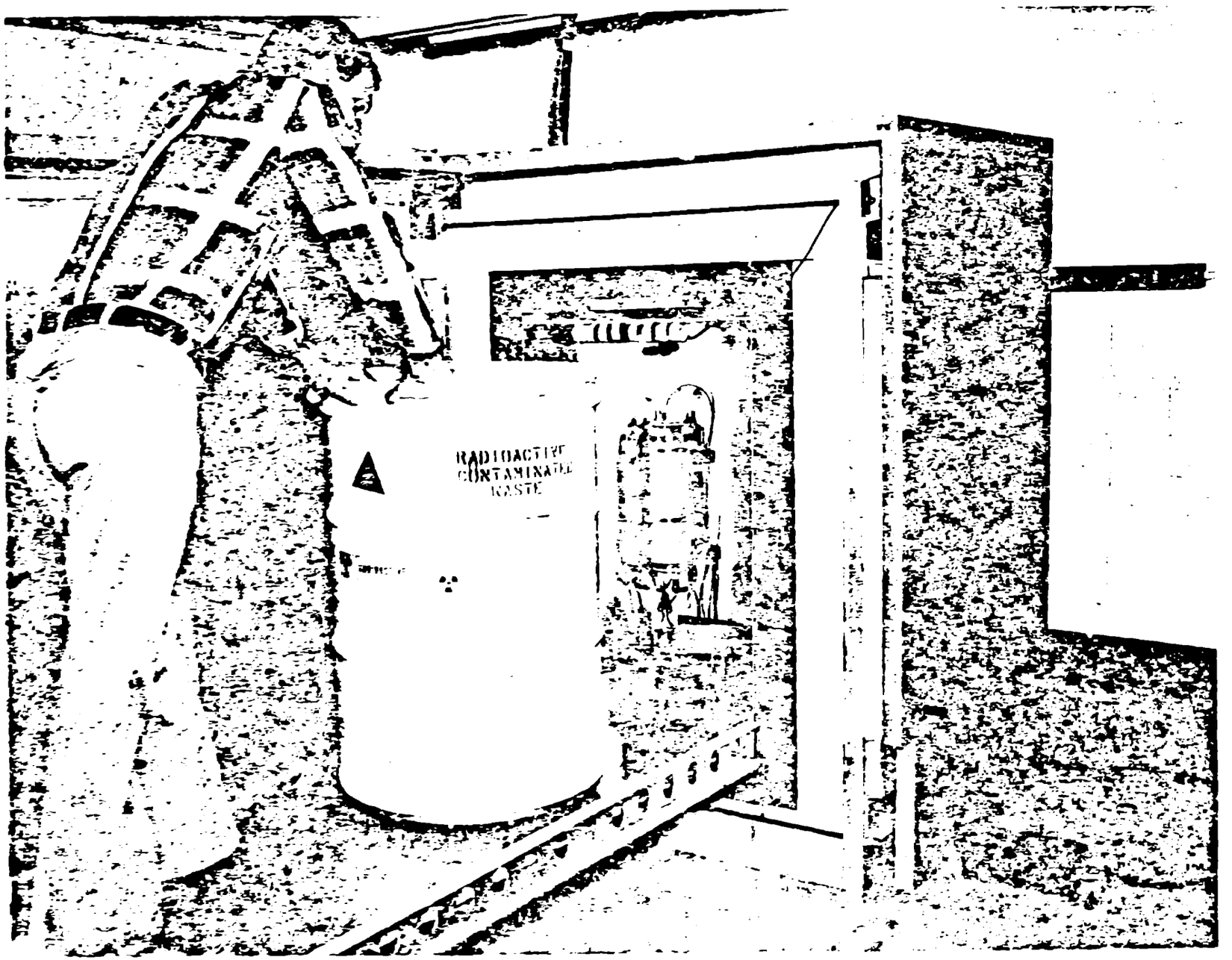
BARREL FILLED WITH
SAND AND VERMICULITE
D - T NEUTRON GENERATOR
45.72 cm x 10.16 cm


RADIOACTIVE
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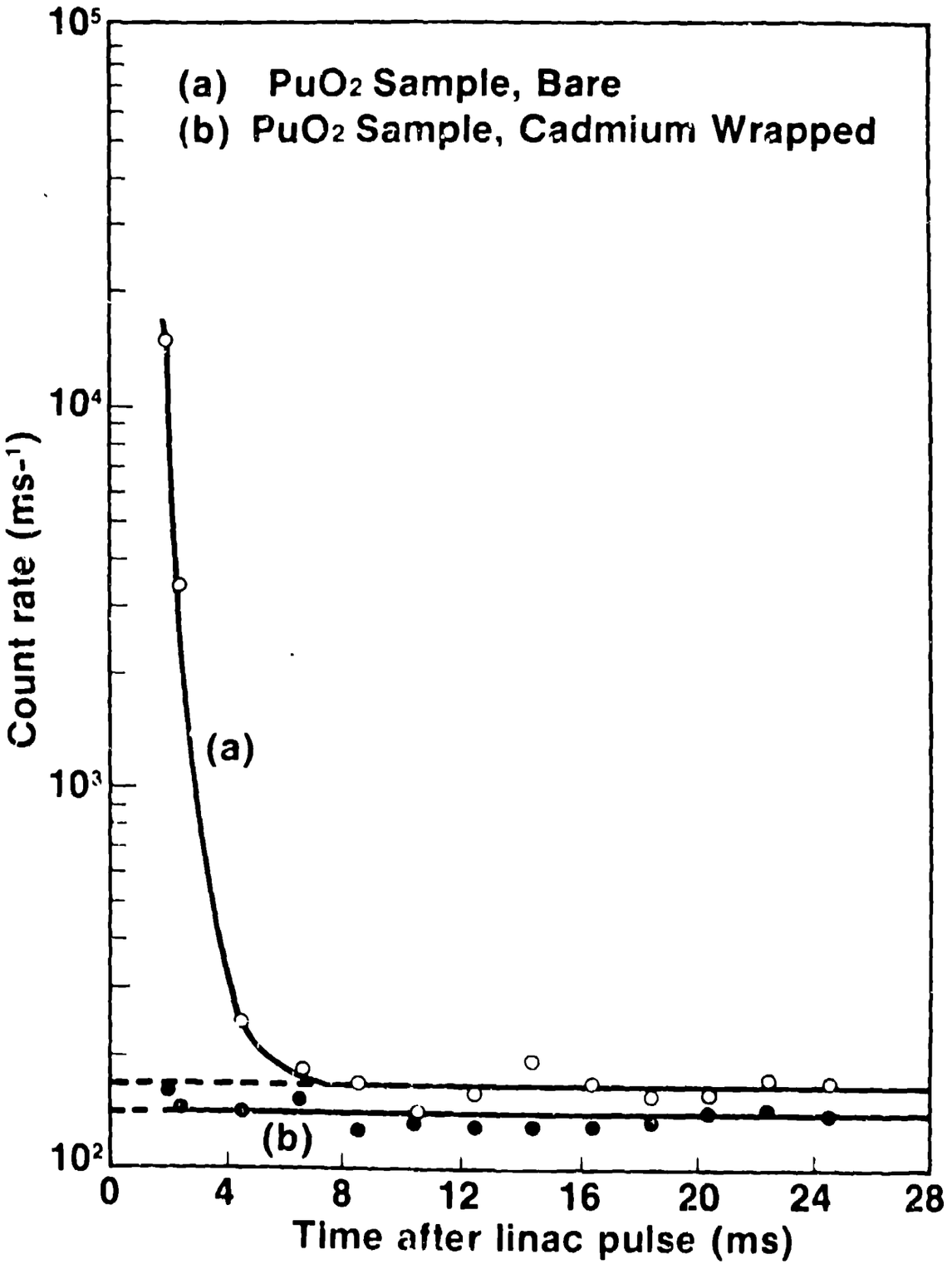
(a) PuO2 Sample, Bare
(b) PuO2 Sample, Cadmium Wrapped
Count rate (ms-1)
10⁵
10⁴
10³
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(a)
(b)
0
4
8
12
16
20
24
28
Time after linac pulse (ms)